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Author(s)	
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Division of Multidisciplinary Chemistry - Molecular Aggregation Analysis -

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Prof
SATO, Naoki
(D Sc)



Assoc Prof
ASAMI, Koji
(D Sc)



Assist Prof
KITA, Yasuo
(D Sc)



Assist Prof
YOSHIDA, Hiroyuki
(D Sc)



Proj Res*
MURDEY, Richard James
(Ph D)

*Assist Prof (SER) of
Pioneering Research Unit
for Next Generation



Res
KATO, Keiichi
(D Sc)

Students

HIRAMATSU, Takaaki (D3)
TSUTSUMI, Jun'ya (D3)
MORIE, Jun (M2)
SASAMURA, Tatsuya (M1)

Scope of Research

The research at this subdivision is devoted to correlation studies on structures and properties of both natural and artificial molecular aggregates from two main standpoints: photoelectric and dielectric properties. The electronic structure of organic thin films is studied using photoemission and inverse photoemission spectroscopies in connection with the former, and its results are applied to create novel molecular systems with characteristic electronic functions. The latter is concerned with heterogeneous structures in microcapsules, block polymers, biological membranes and biological cells, and the nonlinearity in their dielectric properties is also studied in relation to molecular motions.

Research Activities (Year 2006)

Presentations

Dielectric Properties of Biological Cells and Tissues: Monitoring and Modeling, Asami K, International Conference of Biosensing and Biodynamics: From Basics to Applications (Bucharest, Romania), 18–21 May.

Observation of Electronic Structure of Frontier States in Organic Semiconductor Thin Films, Sato N, Yoshida H, Murdey R J, Workshop on Charge Carrier Injection and Transport Phenomena in Organic Thin Films and the 10th European Conference on Organised Films (ECOF-10) (Riga, Latvia), 20–24 August.

Dielectric Properties of Osmotically-lysed Erythrocytes, Asami K, 4th Conference of International Dielectric Society & 9th Conference of Dielectric and Related Phenomena (Poznan, Poland), 3–7 September.

Structure and Electronic Structure of Unoccupied States in Pentacene Thin Films, Sato N, Yoshida H, Murdey R J, The 13th International Conference on Solid Films and Surfaces (ICSFS-13) (San Carlos de Bariloche, Argentina), 6–10 November.

Structure of Pentacene Thin-Film Phase Determined with X-ray Diffraction Reciprocal Space Mapping Method (in Japanese), Yoshida H, Inaba K (Rigaku Co.), Sato N, The 15th Organic Crystals Symposium (Matsuyama, Japan), 24–25 November.

IPES and UPS Studies on Frontier Electronic States in Organic Semiconductor Thin Films, Sato N, Yoshida H, Murdey R J, Asian Pacific Conference on Surface Science & Engineering (Hong Kong, China), 19–21 December.

Grants

Sato N, Development of Novel Electronic Systems Based on Hybridization of Characteristic Molecular Properties and Specific Aggregate Structures, Grant-in-Aid for Scientific Research (2) on Priority Areas of Molecular Conductors, 17 October 2003–31 March 2008.

Asami K, Dielectric Spectroscopy of Blood Cells, Collaboration Research with Sony Corporation (Life Science Laboratory, Material Laboratories), 10 November 2006–31 March 2007.

Polymorphic Phase Transition of the Crystal of a Highly Amphoteric Polar Molecule

{4-[4,5-Bis-(methylsulfanyl)-1,3-dithiol-2-ylidene]-cyclohexa-2,5-dien-1-ylidene}malononitrile (BMDCM) grouped into highly amphoteric polar molecules was found to show polymorphism: Solution- and vapor-grown crystals are blue plate-formed and green needle-formed ones, respectively. X-ray structural analyses revealed that the space groups of the former and the latter crystals are triclinic $P\bar{1}$ and monoclinic $P2_1/n$, respectively. BMDCM molecules are stacked along *a*-axis in the head-to-tail manner to form π electron columns in both crystals. The orientation relation among the stacked molecular columns is different between the two crystals; all the molecular long axes are parallel to each other in the solution-grown crystal, however, the molecular long axes of a column are almost normal to those of the nearest column in the vapor-grown one. Further, the solution-grown crystal exhibited color change from blue to green when heated at 493 K. Then we examined powder X-ray diffraction patterns of the crystal before and after the heat treatment and confirmed that the pattern obtained for the heated crystal was almost the same as that for the vapor-grown crystal. Thus we have found the polymorphic phase transition of BMDCM crystal induced thermally. It is notable that a larger solution-grown crystal showed the phase transition with keeping the crystalline appearance in spite of the large difference in the packing manner of molecular stacking columns between the two polymorphs. Such a difference and the phase transition behavior attract our attention in terms of electronic properties and molecular dynamics in the solid state so that further studies on these problems are in progress.

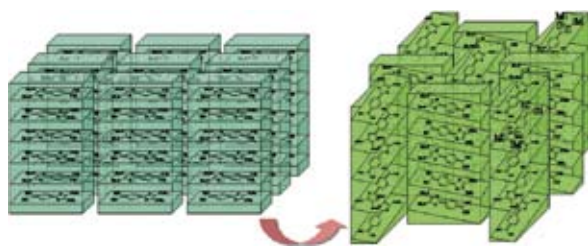


Figure 1. Schematic of the change in packing manner of molecular stacking columns via polymorphic phase transition of BMDCM crystal.

Ion Channels of N-Terminally-Linked Alamethicin Dimers: Enhancement of Cation-Selectivity by Substitution of Glu for Gln at Position 7

Alamethicin forms voltage-gated ion channels that have moderate cation-selectivity. The enhancement of the cation-selectivity by introducing negatively charged residues at positions 7 and 18 has been studied using the tethered homodimers of alamethicin with Q7 and E18 (di-alm-Q7E18) and its analog with E7 and Q18 (di-alm-E7Q18) (Figure 2). In the dimeric peptides, monomer peptides are linked at the N-termini by a disulfide bond. Both the peptides formed long lasting ion channels at *cis*-positive voltages when added to the *cis*-side membrane. Their long open duration enabled us to obtain current-voltage ($I-V_m$) relations and reversal potentials at the single-channel level by applying a voltage ramp during the channel opening. The reversal potentials measured in asymmetric KCl solutions indicated that ionized E7 provided strong cation-selectivity whereas ionized E18 little influenced the charge selectivity. This was also the case for the macroscopic charge selectivity determined from the reversal potentials obtained by the macroscopic $I-V_m$ measurements. The results are accounted for by stronger electrostatic interactions between permeant ions and negatively-charged residues at the narrowest part of the pore than at the pore mouth.

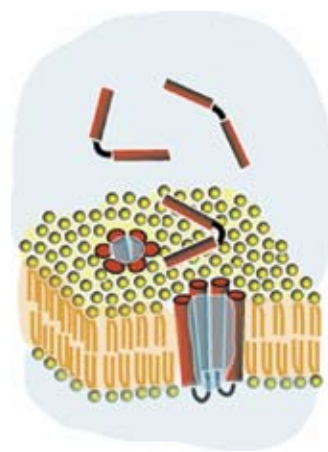


Figure 2. Ion channels formed by covalently-linked alamethicin dimers in a lipid bilayer.

Yoshida H, Controlling Reactivity and Diffusion at Metal-Organic Semiconductor Interfaces through the Deposition of Metal Clusters, Grant-in-Aid for Scientific

Research for Young Scientists (B), 19 October 2004–31 March 2006.